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Analysis of the Variability of Classsified and Unclassified Radiological Source term Inventories in the Frenchman Flat Area, Nevada test Site

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Letter Report

Analysis of the Variability of Classified and Unclassified Radiological Source Term Inventories in the Frenchman Flat Area, Nevada Test Site

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It has been proposed that unclassified source terms used in the reactive transport modeling investigations at NTS CAUs should be based on yield-weighted source terms calculated using the average source term from Bowen et al. (2001) and the unclassified announced yields reported in DOE/NV-209. This unclassified inventory is likely to be used in unclassified contaminant boundary calculations and is, thus, relevant to compare to the classified inventory. We have examined the classified radionuclide inventory produced by 10 underground nuclear tests conducted in the Frenchman Flat (FF) area of the Nevada Test Site. Our goals were to (1) evaluate the variability in classified radiological source terms among the 10 tests and (2) compare that variability and inventory uncertainties to an average unclassified inventory (e.g. Bowen 2001).

To evaluate source term variability among the 10 tests, radiological inventories were compared on two relative scales: geometric mean and yield-weighted geometric mean. Furthermore, radiological inventories were either decay corrected to a common date (9/23/1992) or the time zero (t_0) of each test. Thus, a total of four data sets were produced. The date of 9/23/1992 was chosen based on the date of the last underground nuclear test at the Nevada Test Site. The geometric mean activity for each radionuclide was calculated as follows:

$$GM_{RN} = \sqrt[n]{\prod_i A_{RN,i}} \qquad (1)$$

where $A_{RN,i}$ is the activity (A) for a particular radionuclide (RN) for test i and n is the total number of tests. A yield-weighted geometric mean activity was calculated as follows:

$$GM_{RN,Y} = \sqrt[n]{\prod_i \frac{A_{RN,i}}{Y_i}} \qquad (2)$$

where Y_i is the classified yield of test i. The relative inventory for each radionuclide and each test was calculated by dividing the test-specific radionuclide activity (or yield-weighted activity) by the associated geometric mean activity:

$$\log Ratio_{RN,i} = \log\left(\frac{A_{RN,i}}{GM_{RN}}\right) = \log A_{RN,i} - \log GM_{RN} \qquad (3)$$

$$Standard\ Deviation\ of\ log(Ratio_{RN,i}) = \sqrt{\frac{\sum(\log Ratio_{RN,i} - \overline{\log Ratio_{RN,i}})^2}{n-1}} \quad (4)$$

or the yield-weighted geometric mean activity:

$$\log Ratio_{RN,Y,i} = \log(\frac{A_{RN,i}/Y_i}{GM_{RN,Y}}) = \log(A_{RN,i}/Y_i) - \log GM_{RN,Y} \quad (5)$$

$$Standard\ Deviation\ of\ log(Ratio_{RN,Y,i}) = \sqrt{\frac{\sum(\log Ratio_{RN,y,i} - \overline{\log Ratio_{RN,Y,i}})^2}{n-1}} \quad (6)$$

As stated earlier, relative inventories and yield-weighted relative inventories were calculated at a single date (9/23/1992) and at the t_0 for each test. This analysis generated four datasets (1992, t_0, yield weighted1992, yield weighted t_0) which could be discussed in an unclassified format because classified test-specific source term information is not available from the relative scale employed here.

All calculations were performed on a total of 33 radionuclides. Some 9 radionuclides including 3 activation products ^{26}Al, ^{93m}Nb, ^{150}Eu, and 6 actinides ^{232}U, ^{233}U, ^{236}U, ^{237}Np, ^{243}Am and ^{244}Cm, were excluded due to data inadequacies. ^{40}K, as a natural radionuclide, was also excluded. The activities of the excluded radionuclides are negligible comparing to the total inventory in FF CAU. The removal of these radionuclides allowed us to focus on those radionuclides that are more relevant to modeling. For radionuclides that included both natural and device sources (^{232}Th, ^{235}U, and ^{238}U), the combined natural + device activity was used in this analysis. The relative activity results (Figure 1) suggest that radionuclide activities for individual tests typically fall within a little less than 2 order of magnitude ranges at FF except the activation products. This variability is substantially larger than the inventory uncertainties reported in Bowen et al. (2001, see Table 1) and may reflect differences in test yield, performance, and individual radionuclide, etc.

Table 1. Estimated classified inventory accuracies for various groups of radionuclides. From Bowen et al. (2001).

	% Error	Order of magnitude
Fission Products	~10 to 30%	0.04-0.11
Unspent Fuel	20% or better	< 0.08
Fuel Activation Products	50% or better	< 0.18
Residual Tritium	300% or better	< 0.48
Activation Products	A factor of 10	1

Similar to the observations in data analyses for RM/SM area (Zhao and Zavarin, 2008), that the variability of the relative activity in fission product source term is reduced significantly

when weighted by yield. Figure 2 shows yield-weighted radionuclide activities relative to yield-weighed geometric mean decay corrected to September 23, 1992. Similar plots were obtained for yield weighted relative activities at t_0, however, the plots are not shown in this report. It should not be surprising given that the production of fission products should be strongly correlated to the quantity of fissioned device fuel. Furthermore, uncertainty in the classified source terms is relatively small (~10 to 30%, as reported in Bowen et al., 2001) when compared to the range of test yields at FF. Thus, the relationship between yield and fission product activity is clearly discernible. Variability in activation products almost remains the same. Both ranges of tritium and actinide relative activity increase 0.5 log unit when weighted by yield. For actinides, the residual actinide activity may, in fact, be negatively correlated with yield since efficient and large detonations might result in very little residual fuel.

To facilitate further interpretation of source term variability, we partitioned the 33 radionuclides into 4 groups. The four groups were based on their primary source, as defined in Bowen et al. (2001): Tritium, activation products, fission products, and actinides. The 4 groups are the following:

1. Tritium: ^{3}H
2. Activation Products: ^{14}C, ^{36}Cl, ^{39}Ar, ^{41}Ca, ^{59}Ni, ^{63}Ni, ^{94g}Nb, ^{152}Eu, ^{154}Eu and ^{166m}Ho, total 10 radionuclides.
3. Fission Products: ^{85g}Kr, ^{90}Sr, ^{93}Zr, ^{99}Tc, ^{107g}Pd, ^{113m}Cd, ^{121m}Sn, ^{126}Sn, ^{129}I, ^{135g}Cs, ^{137}Cs, and ^{151}Sm, total 12 radionuclides.
4. Actinides: ^{232}Th, ^{234}U, ^{235}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{241}Am, total 10 radionuclides.

Some radionuclides are products of both activation and fission. We assigned these radionuclides according to their primary source. A relative activity range and standard deviation was calculated for each of the 4 radionuclide groups. Table 2 summarizes the relative activity ranges and standard deviations for each radionuclide group for all tests at Frenchman Flat. With a decrease in the number of tests in FF CAU, the relative activity ranges and the standard deviations decreased as well comparing to the same set of data for RM/SM CAU.

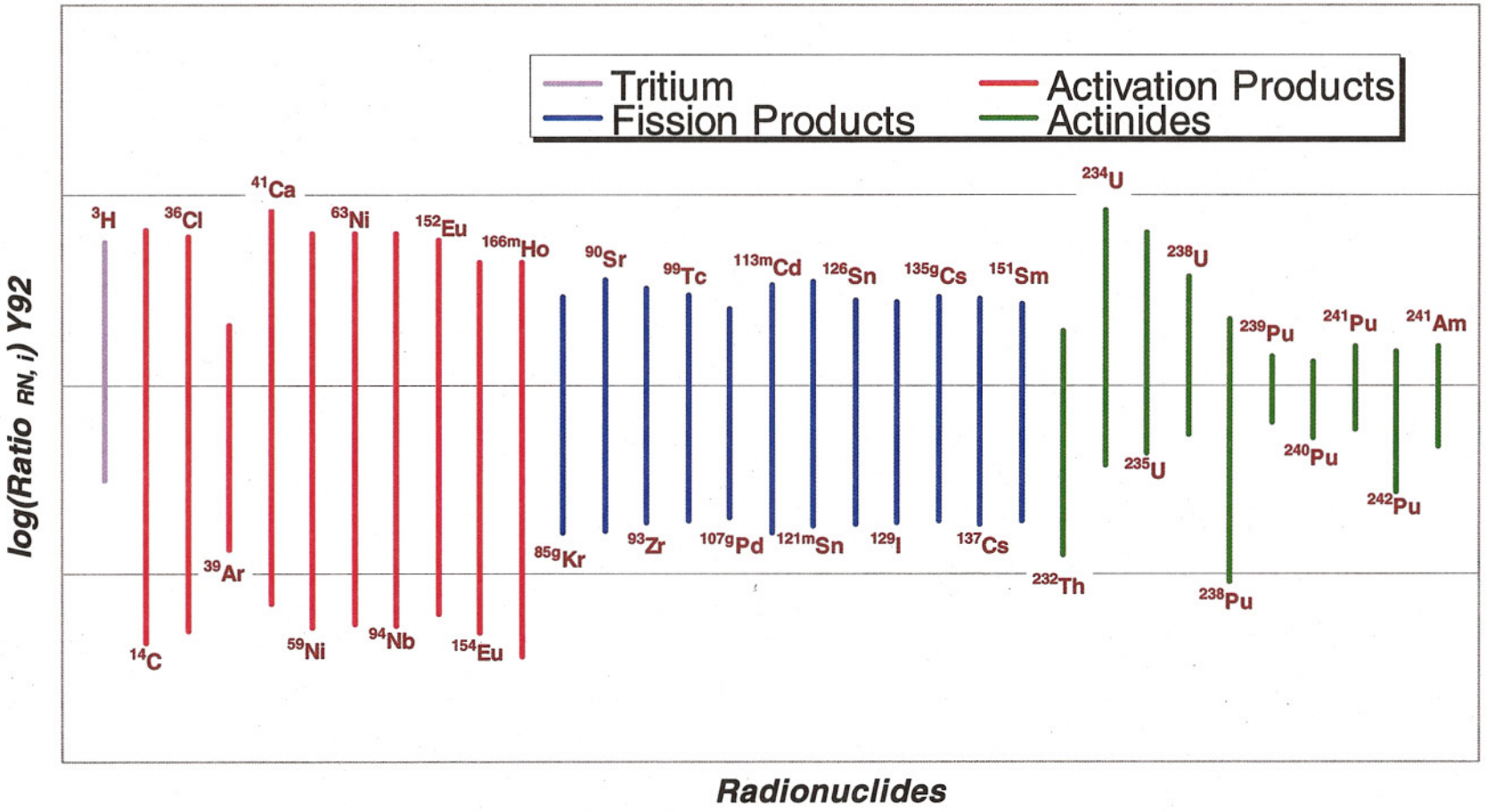

Figure 1. Ranges of radionuclide activities relative to geometric mean decay corrected to September 23, 1992.

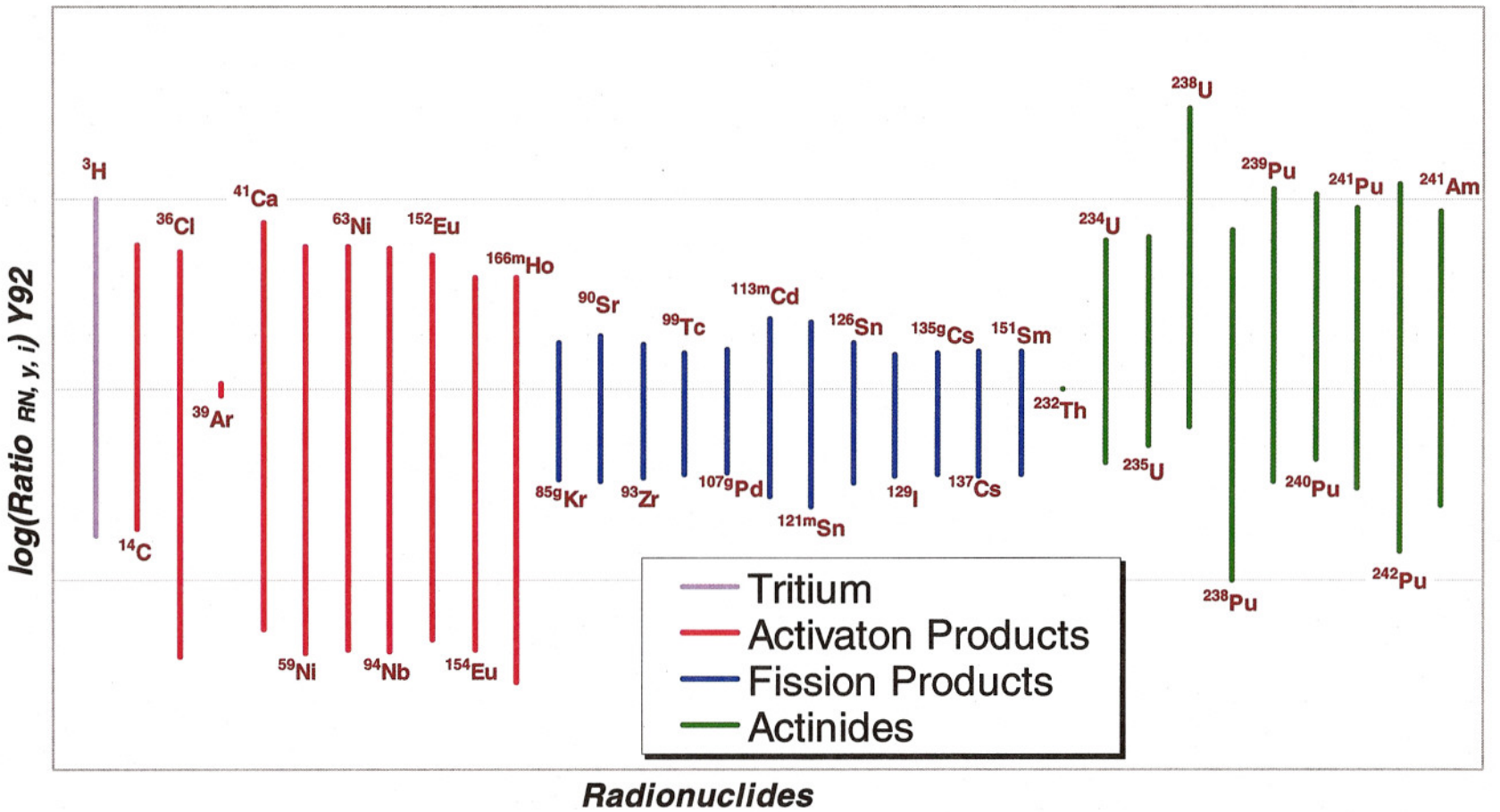

Figure 2. Ranges of yield-weighted radionuclide activities relative to yield-weighed geometric mean decay corrected to September 23, 1992.

Table 2. Range of Radionuclide Inventory at Frenchman Flat

	Range of $Log(Ratio_{RN,i})$ (Y92)	Standard Deviation of log unit	Range of $Log(Ratio_{RN,Y,i})$ (Y92)	Standard Deviation of log unit
Tritium	1.26	0.47	1.77	0.63
Activation Products	2.37	0.68	2.41	0.56
Fission Products	1.34	0.40	0.98	0.27
Actinides	1.97	0.29	2.48	0.40

units are order of magnitude

In general, yield weighting increases the range of tritium activity and the standard deviation by 0.5 and 0.2 log unit, respectively. The variability is at a scale similar to the tritium inventory accuracy reported by Bowen et al. (2001) and, thus, may simply reflect source term uncertainty. Yield-weighting of activation products results in minimum changes in both their range and standard deviation. However, this may simply result from the small number of tests detonated in FF CAU. As expected, yield weighting the fission products reduces relative activity range and the standard deviations by 0.4 and 0.1 log units, respectively. The activities of fission products are clearly proportional to the reported test yields. The remaining variability can, most likely, be attributed to source term uncertainties (Table 1). Yield weighting of actinides increases the actinide relative activity range and the standard deviation by 0.5 and 0.1 log units. For the tests with similar amount of actinide fuel, the test with higher yields may result in smaller actinide residual source terms because they effectively burn their actinide fuels. The exception in the actinide group is ^{232}Th, whose variability decreased drastically after yield-weighting, suggesting that the ^{232}Th inventory is predominantly of natural origin.

As general guidelines, yield-weighting of fission product radionuclides addresses a large proportion of the source term variability in FF tests. The remaining variability can, most likely, be attributed to uncertainties in the classified inventories, as defined in Table 1. The yield-weighting addresses very little or none of the variability in tritium, activation products and actinide source terms. Thus, yield weighting is not an appropriate means for addressing variability of these source terms at FF. Importantly, the yields used here were classified. Conducting this analysis using announced unclassified yields (i.e. DOE/NV-209) would substantially increase the variability in all yield-weighted calculations (see discussion below).

Histograms of radionuclide relative activity on reference date of 9/23/1992 are plotted in Figures 3 and 4. In most cases, the histograms show that activity distributions are asymmetric, especially, for groups of activation and fission products. The comparison between Figure 3 and Figure 4 indicates that the yield-weighting fission product substantially narrows the range of their relative activities. This suggests that yield weighting addresses most of the source term variability for fission products. For the other radionuclides, including tritium, activation products and actinides, yield-weighting increases the variability. Therefore, yield-weighting these radionuclides is not recommended.

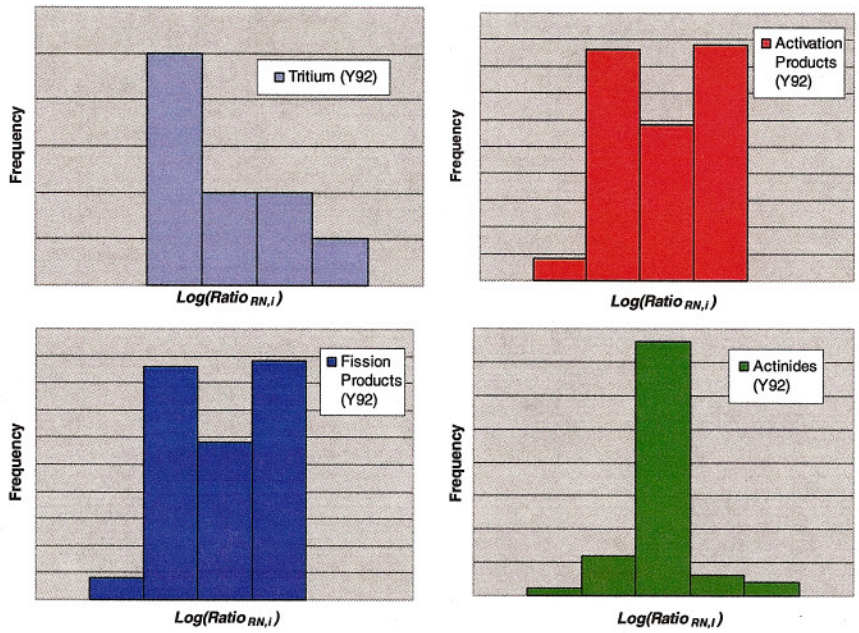

Figure 3. Histograms of radionuclide activities relative to their geometric means decay corrected to 9/23/1992.

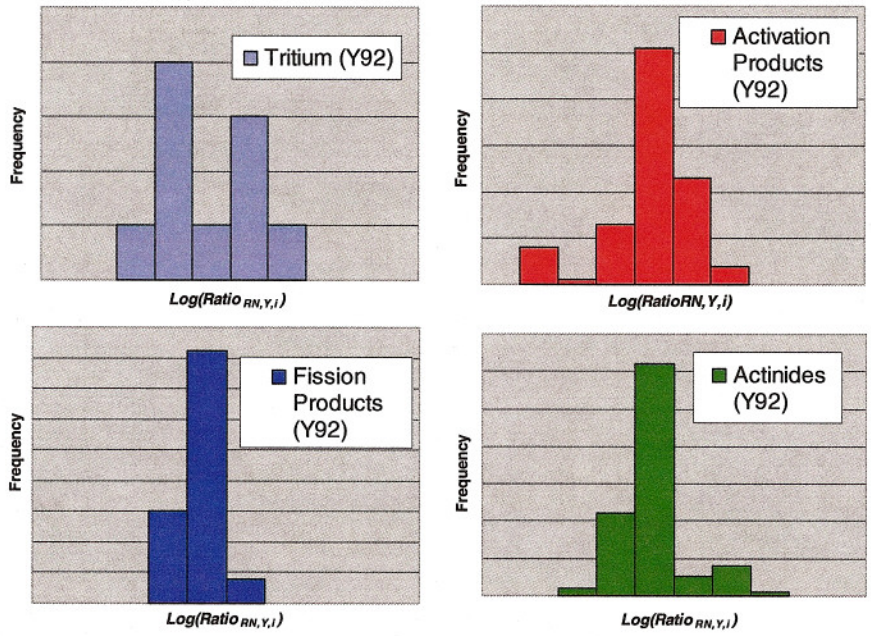

Figure 4. Histograms of yield-weighted radionuclide activities relative to their yield-weighted geometric means decay corrected to 9/23/1992.

It has been proposed that unclassified source terms used in reactive transport modeling investigations should be based on yield-weighted source terms calculated using the average source term from Bowen et al. (2001) and the unclassified announced yields reported in DOE/NV-209. This unclassified inventory is likely to be used in unclassified contaminant boundary calculations and is, thus, relevant to compare to the classified inventory. Many of the announced yields reported in DOE/NV-209 are, in fact, ranges. Therefore, the maximum yield of the range was used here to calculate the unclassified yield-weighted inventory for each of the 33 radionuclides in each test.

The unclassified yield-weighted inventory of all FF tests and all 33 radionuclides examined previously was compared to the classified inventory. Table 3 summarizes 95% confidence intervals for correlation of unclassified yield-weighted activities and their classified counterpart for tests in Frenchman Flat area (in log unit). Importantly, most classified radionuclide source terms fall within an order of magnitude of the unclassified yield-weighted average. This is encouraging that it suggests that unclassified contaminant boundary calculations are not likely to differ dramatically from their classified counterparts.

Table 3. 95% Confidence Intervals for difference between classified activities and unclassified yield-weighted activities for tests in FF area (log unit).

# of tests	Tritium	Activation Products	Fission Products	Actinides
10 tests	1.28	1.15	0.80	1.06

The confidence intervals for correlations between unclassified yield-weighted inventories and classified inventories are plotted for tritium, activation products, fission products, and actinides in Figures 5 through 8. The lines represent 68% (red) and 95% (blue) confidence intervals (CI), respectively, for each radionuclide group. The unclassified yield-weighted inventory is calculated for each test and each radionuclide using the average Frenchman Flat source term (Bowen et al., 2001) and the maximum announced yield (DOE/NV-209):

$$A_{U,RN,i} = \frac{A_{U,RN,T}}{Y_{U,T}} Y_{U,i} \qquad (7)$$

Where $A_{U,RN,T}$ is the total Frenchman Flat activity for a radionuclide (Bowen et al., 2001), $Y_{U,T}$ is the total unclassified yield based on maximum announced yields (DOE/NV-209), and $Y_{U,i}$ is the unclassified maximum announced yield for test i.

The confidence intervals are calculated from the log-scale differences between unclassified yield weighted inventories and classified inventories for all ten tests ($\log A_{RN,i} - \log A_{U,RN,i}$). Based on this analysis, it is apparent that most unclassified

radionuclide inventories fall within an order of magnitude of their classified counterparts (Table 3). Not surprisingly, the confidence interval for fission products is the lowest of all radionuclide groups. The 95% confidence interval for activation products is at the same scale as the classified inventory accuracy (Table 1) estimated by Bowen et al. (2001) while that of tritium, fission products, and actinides is substantially larger.

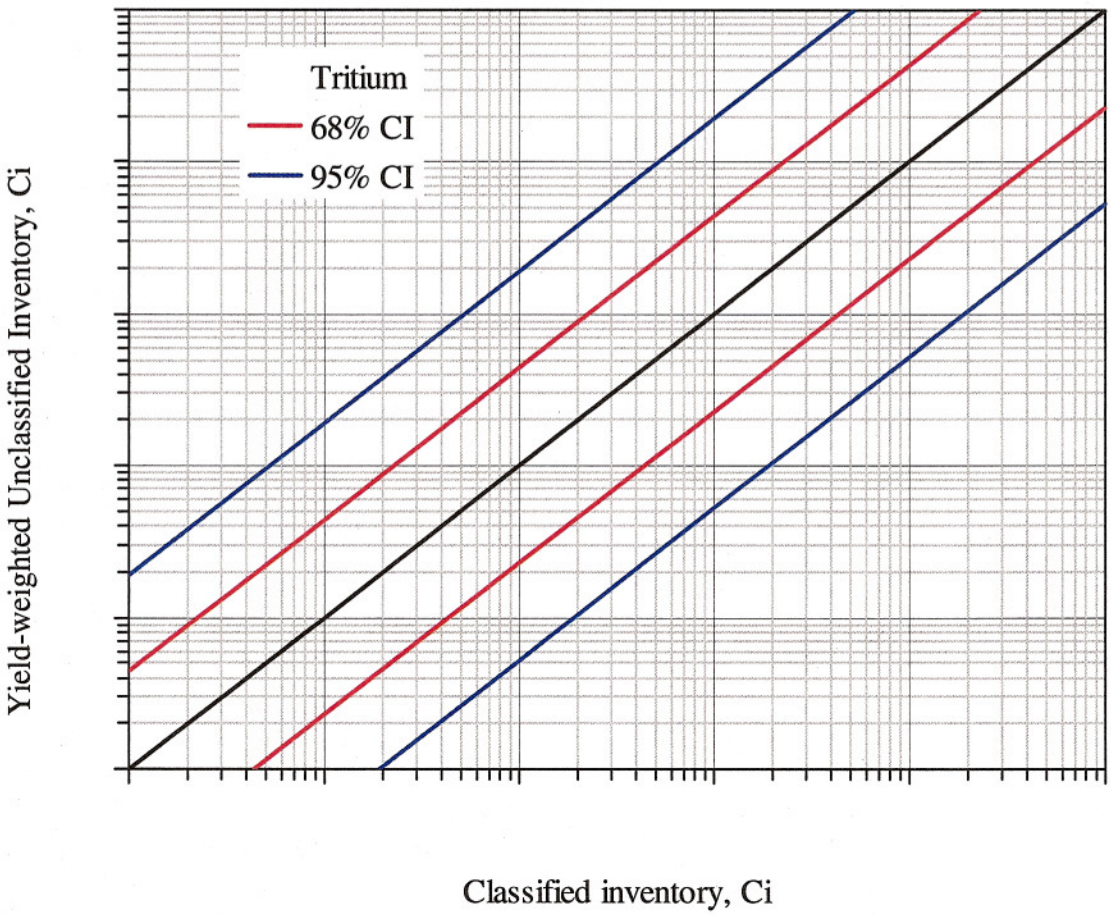

Figure 5. Tritium source term correlation between classified inventories and yield-weighted unclassified inventories for tests detonated in Frenchman Flat CAU. Red lines and blue lines represent 68% and 95% confidence intervals (CI), respectively.

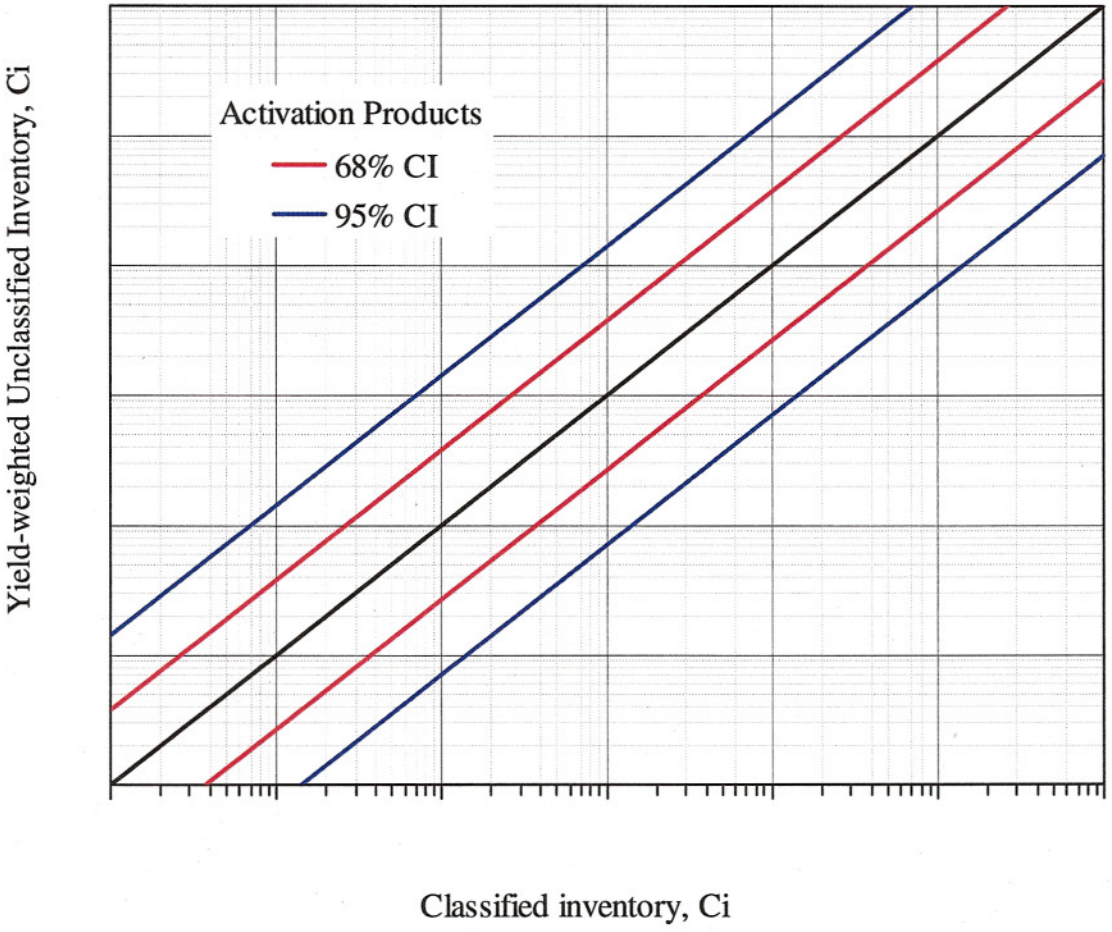

Figure 6. Activation product source terms correlation between classified inventories and yield-weighted unclassified inventories for tests detonated in Frenchman Flat CAU. Red lines and blue lines represent 68% and 95% confidence intervals, respectively.

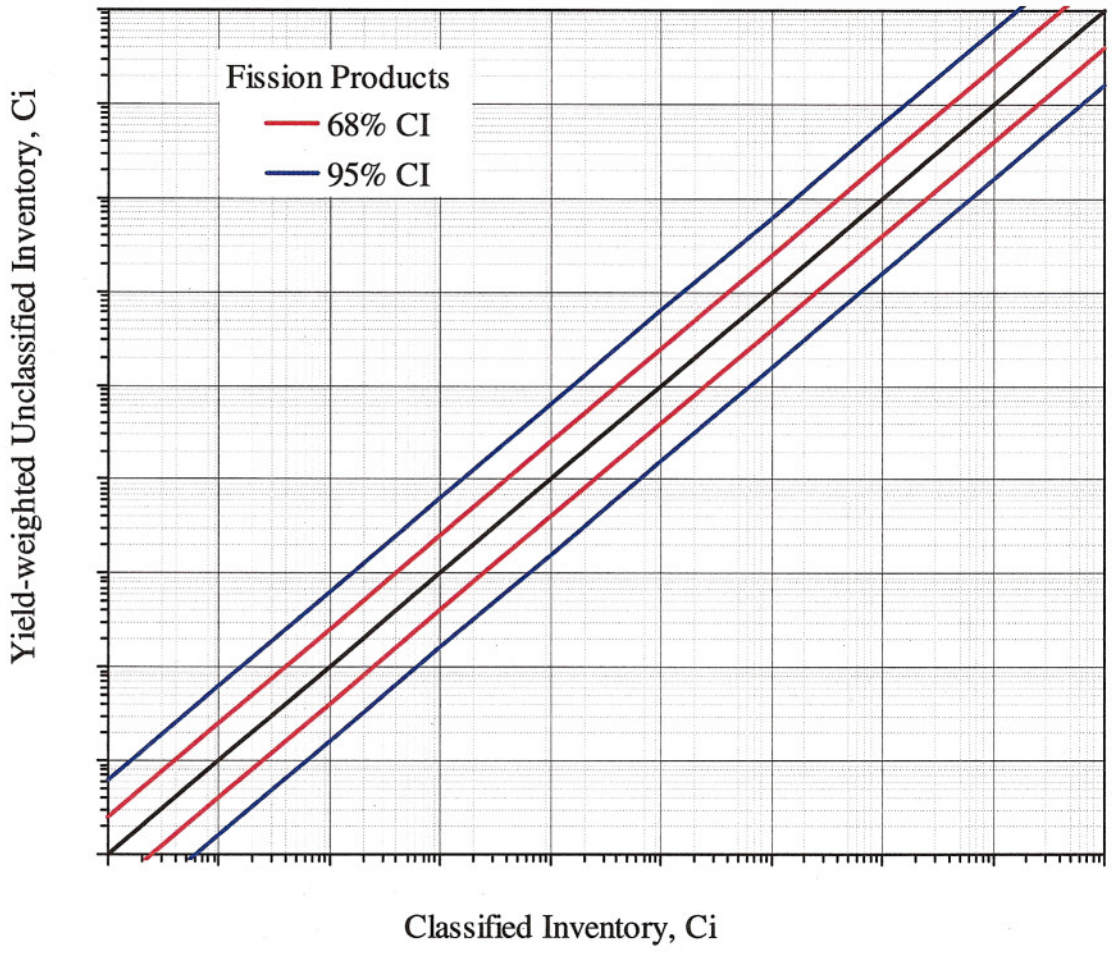

Figure 7. Fission product source terms correlation between classified inventories and yield-weighted unclassified inventories for tests detonated in Frenchman Flat CAU. Red lines and blue lines represent 68% and 95% confidence intervals, respectively.

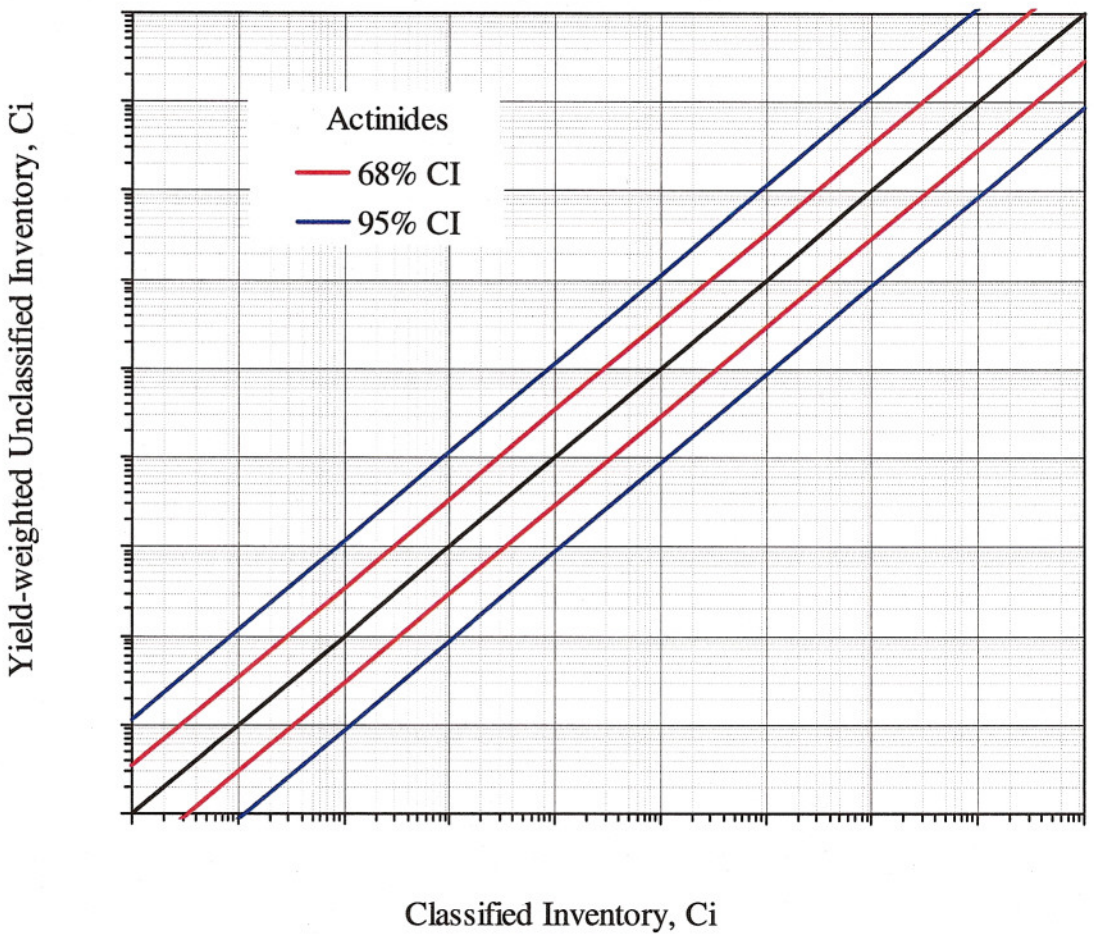

Figure 8. Actinide source term correlation between classified inventories and yield-weighted unclassified inventories for tests detonated in Frenchman Flat CAU. Red lines and blue lines represent 68% and 95% confidence intervals, respectively.

CONCLUSIONS

Based on our analysis of yield-weighted classified inventories, we find that much of the variability in fission product inventories can be attributed to test yield. The remaining variability can be attributed to the uncertainty in classified source term estimates (i.e. Table 1). However, actinide fuel variability cannot be attributed to yield. This is likely because efficient high-yield tests would tend to burn up most of their fuel, resulting in low actinide concentrations. Conversely, tests that did not perform to specifications may have low yields and significant amounts of unburned fuel. Tritium and activation product source terms do not correlate very strongly with yield, in part due the large uncertainties associated with its classified inventory and its different uses and sources on various tests.

When comparing classified source terms to the unclassified yield-weighted source terms calculated using information from Bowen et al. (2001) and DOE/NV-209, we find that

a majority of unclassified radionuclide inventories fall within one order of magnitude of the classified inventory. This is encouraging in that it suggests that unclassified contaminant boundary calculations are not likely to differ dramatically from their classified counterparts.

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